From *Marine Pollution Bulletin* (1998) vol. 37: 14-19

Mussel Watch Results from 1986 to 1996

Thomas P. O'Connor
NOAA N/ORCA21
National Status and Trends Program
1305 East West Highway
Silver Spring, MD 20910

Abstract

Mussel Watch results from coastal waters of the United States for 1986 through 1996 show decreasing trends in molluscan concentrations of chlorinated hydrocarbons, butyltins, and cadmium. Weaker cases for decreases exist for copper , zinc, and silver. Concentrations of polycyclic aromatic hydrocarbons, arsenic, mercury, nickel, lead, and selenium show no overall trend.

Introduction

In 1986 the Mussel Watch Project within the National Status and Trends (NS&T) Program of the U. S. National Oceanic and Atmospheric Administration began to monitor spatial distributions and temporal trends of contaminant concentrations in coastal and estuarine regions of the United States. Results up to 1993 from annual collections and analyses of mussels and oysters (O'Connor and Beliaeff 1995; O'Connor ,1996; Beliaeff *et al* .,1997) indicated that the general trend was a temporal decrease involving mainly chlorinated hydrocarbons and butyltin, whose use has been banned; and cadmium, whose use has been greatly curtailed. Results through 1996 remain essentially the same.

Site Locations

Lauenstein *et al.* (1997) provided detailed descriptions of each Mussel Watch sampling site. Distributed throughout the marine coastal United States, they are on average 20 km apart in estuaries and embayments and 100 km apart along open coastlines. They have been selected with the intention of collecting samples that are "representative" of their surroundings. Small scale patches of contamination and known points of waste discharge have been avoided. A file of site locations accompanies the chemical data file that is available on the internet at http://seaserver.nos.noaa.gov/projects/nsandt/nsandt.html

Since 1986 and through 1996, the Mussel Watch project has sampled 287 sites. Of those, 196 have been sites occupied in at least six years and data from them have been examined for temporal trends. The other 83 sites include those recently added to the program, such as sites in the Great Lakes, and those sampled only once or twice to temporarily increase spatial resolution in specific areas. Since 1994 the Mussel Watch program has begun to sample sites on a biennial basis. Sampling occurs every year but not at all sites.

Chemicals Monitored

The elements and compounds measured in the NS&T Program are listed in Table 1. Except for Al, Fe, and Mn, the elements in Table 1 are all possible contaminants in the sense that their concentrations in the environment have been altered by human activities (Nriagu, 1989). The mere existence of the chlorinated organic compounds and butyltins indicates human activity. Polycyclic aromatic hydrocarbons (PAHs) are similar to metals in the sense that they occur naturally. They are found in fossil fuels such as coal and oil and are produced during the combustion of organic matter. Their environmental presence is also attributable to humans because they are released in the use and transportation of petroleum products and from a multitude of human activities, such as burning coal and wood or incinerating waste. Almost all the chemicals in Table 1 are also on the list of 127 Priority Pollutants created by the United States Environmental Protection Agency in the late 1970s (Keith and Teillard, 1979).

Seven PAH compounds— acenaphthylene, 1,6,7-trimethylnaphthalene, benzo[*b*]- and benzo[*k*]fluoranthene, indeno[1,2,3-*cd*]pyrene, and benzo[*ghi*]perylene— were not measured until 1988. In this paper, trends for PAHs will be examined for 1988 to 1993. Also in 1988, the program began measuring the 18 PCB congeners listed above, while in 1986 and 1987 PCBs were quantified to level of chlorination. To maintain consistency over the years for trend analysis, the post-1987 total PCB concentrations have been adjusted as follows:

tpcb=tpcbcon*1.945+3.35 for data from Battelle
tpcb=tpcbcon*2.165+2.82 for data from Texas A&M/GERG

where tpcb is the sum of PCBs at each level of chlorination and tpcbcon is the sum of 18 congeners. These regressions are based on 150 analyses by each of two laboratories in 1987 where PCBs were quantified both to level of chlorination and by congener. In each case $r^2 \geq 0.96$. If no congeners were detected in a sample (i.e. tpcbcon=0), tpcb was also set to zero. Lastly, systematic monitoring of tributyltin and its metabolites di- and mono-butyl tin did not begin until 1989, so trends can only be examined from that year.

Methods

Since 1986 all samples from the Gulf of Mexico Coast and since 1995 all samples from all coasts have been collected and analyzed by scientists from the Geochemical and Environmental Research Group or the Department of Oceanography at Texas A&M University in College Station, TX (GERG/TAMU). For 1986 through 1988, samples from California were collected and analyzed by Scientific Applications International Corporation (SAIC) in LaJolla, CA. Between 1986 and 1994, all other West Coast samples and all East Coast samples were collected and analyzed by scientists from the Battelle Laboratories in Duxbury,

MA and Sequim, WA. Detailed descriptions of sampling and analytical protocols are available (Lauenstein and Cantillo, 1993a, b, c, and d).

Earlier analyses of trends in the NOAA data (O'Connor and Beliaeff 1995; O'Connor ,1996; Beliaeff *et al* .,1997) did not include results for silver or for chromium because the analytical data were being reassessed. No errors have been in the original chromium analyses. The original digestions for silver were incomplete and that led to a reanalysis of all samples for silver (Daskalakis et al, 1997). Samples from the East and West coast were reanalyzed by GFAA following digestion with excess chloride as stipulated by Daskalakis et al. (1997). Samples from the Gulf of Mexico were reanalyzed by INAA. Since 1995 all samples have been analyzed by TAMU using INAA.

Mussels are collected along the entire West Coast and north of Delaware Bay along the East Coast. Oysters are collected at the remaining East Coast sites (Delaware Bay and south), at Gulf Coast sites, and at three sites in Hawaii. Two species of mussels and two species of oysters are collected: the blue mussel *Mytilus edulis* on the East Coast; *M. edulis* and *M. californianus* on the West Coast; the oyster *Crassostrea virginica* in the continental United States; and the oyster *Ostrea sandvicensis* in Hawaii. The preferred size ranges are 5 to 8 cm for mussels, 7 to 10 cm for C*. virginica* and 2.5 to 5 cm for *O. sandvicensis.* Mollusks are not shucked in the field. They are separated when found to be adhering to one another and scrubbed with a nylon or natural fiber brush to remove adhering detritus. Cleaned samples are then packed in dry ice and shipped to the laboratory. Composite samples are prepared by homogenizing the soft parts of 30 mussels or 20 oysters. Six composites are used for chemical analysis, three for replicate organic contaminant analyses and three for trace elements. At all laboratories, composites were homogenized prior to analyses and, at Battelle and Texas A&M, homogenized samples were subsequently freeze-dried. Since 1992, and in all years for butyltins, only one composite sample per site has been collected. Mollusks are collected between November and March with the collection date at each site within three weeks of a specified date.

Lowest reported concentrations

Detection limits and their method of determination varied among years. In this summary all analytical results showing no usable signal were treated as concentrations of zero. This can complicate trend detection because zeros will always be the lowest possible concentration, while non-zeros in other years could possibly indicate greater analytical sensitivity rather than higher concentrations. For the trace elements and the aggregated organic compounds listed above, zeros are not sufficiently common to affect trend analysis. Among 1669 sets of mean concentrations for 1986 through 1996, there are fewer than 9 zeros for all elements other than Ag and Cr where there are 93 and 41 zeros, respectively. . Among the aggregate groups of chlorinated organic compounds there are 118, 37, 13, and 8 zeros for ∑Dieldrin, ∑Cdane, ∑PCB, and ∑DDT, respectively. Among 1381 mean ∑PAH concentrations since 1988, 21 are zero.

Among 1223 ∑BT values since 1989, 140 are zero (45 in 1993 alone). This paucity of non-detectable concentrations for the aggregate groupings of organic compounds does not mean that each individual compound was always detected, but shows that except in a few instances at least one of the constituent compounds in the group was present at a measurable concentration. Table 2 lists the lowest of the non-zero mean concentrations for each element and compound group.

On the other hand, the following chemicals are so infrequently detected that no calculations of trends were made: Sn (29% frequency of no detectable concentration); lindane (19%); mirex (52%); endrin (40%); and hexachlorobenzene (49%). Two trace chemicals, Sb and Tl, were so infrequently detected in 1986 and 1987 that they were not measured after 1987.

RESULTS

Adjustment to change

A central premise for monitoring temporal trends using biological samples is that chemical concentrations in the organism depend on the concentrations in the water and food processed by that organism. Experimental evidence that this is the case for mollusks comes from laboratory experiments where conditions of exposure are controlled (e.g.., Cunningham and Tripp, 1975; Pruell *et al.*, 1987; Fisher and Teyssie, 1986) and from field experiments where oysters or mussels are transplanted from areas of low contaminant concentrations to areas of high concentration and vice versa (Roesijadi *et al.*,1984; Martin, 1985; Capuzzo *et al.*, 1989; Sericano, 1993). There are, nevertheless, a myriad of other factors influencing concentrations. Phillips (1980) and Phillips and Segar (1986) discuss organism size, location relative to tide, salinity, temperature, and reproductive state as factors that can affect molluscan concentrations of chemicals. The NOAA NS&T Program attempts to limit these influences by annually collecting organisms within a set size range, at the same site, and in the winter prior to the spawning season. However, it is not possible to control all these variables and, in effect, they appear as noise in the temporal sequence of concentrations.

Trends

A trend is a statistically significant correlation between contaminant concentration and year. Two non-paramteric tests have been used; Spearman correlations based on rankings of concentrations and Kendall-tau correlations based on directions of change between successive years. Unlike parametric correlations, they are free of assumptions about concentrations at a site being normally distributed with a common variance among sites. All calculations were performed with the SYSTAT package (Wilkinson, 1987).

The results in Table 3 are very similar for both statistical tests and the most common result is a lack of trends. Among the 2744 combinations of 14 chemicals, excluding Ag and Cr, at 196 sites there are 348 decreases and only

88 increases and at the 95% level of confidence (or 73 and 331 using Kendall-tau result). Given a 5% probability of random data showing trends, there could be 69 increases and 69 decreases that are not real trends. Conceivably, none of the increases are real. The important point, however, is that decreases greatly outnumber increases. On a chemical-by-chemical basis, decreases always exceed increases and do so by a factor of three or more for all the chlorinated hydrocarbons, tributyl tin, and Cd. (While not listed here, the results of trend analysis by site and by chemical are available from the author and on the internet via the NOAA State of the Coast Home Page that includes a section on chemical contamination in mollusks <http://state_of_coast.noaa.gov/bulletins/html/ccom_05/ccom.html>.)

Another examination of national trends is based on medians of all data on an annual basis. When calculating medians it is important to recognize species differences. Data from sites in Long Island Sound where both mussels and oysters could be collected (O'Connor, 1994) show that Ag, Cu and Zn are enriched in oysters by more than a factor of 10 relative to mussels, so medians for those elements have been calculated separately in mussels and oysters. Annual national medians are listed in Table 4 along with the Spearman coefficients for their correlation with year.

Except for ∑PCB where the coefficient was slightly less than significant, the same chemicals showed a preponderance of decreases over increases in Table 3. There are also decreasing national medians for copper and zinc in mussels but not in oysters. Due to natural factors both elements are much more concentrated in oysters than in mussels and those same factors may overwhelm any tendency for oysters to reveal trends in environmental levels of either trace element. Trends, on the other hand, could be revealed in mussels. Among the 96 sites where mussels were sampled, the numbers of increasing/decreasing trends were 1/16 for copper and 2/11 for zinc, using the Spearman-rank correlation.

Apparent trends for silver and chromium

Silver concentrations (Table 3) showed a decreasing trend ($p<0.5$) at 24 sites and an increasing trend at one. Conversely, chromium showed an decrease at only 1 site and an increase at 37. In both cases, results are strongly influenced by changes in analytical technique.

Silver was not detected in more than half of the mussels collected in 1995 or 1996. Silver was usually detected in earlier years when the analysis was by GFAA of tissue extracts. Since 1995, however, all silver analysis has been by INAA. This avoids the problem of having to prepare extracts with excess chloride (Daskalakis et al. 1997) that can be used only for Ag analysis, but it is a less sensitive technique. Oysters generally contain ten times more Ag than mussels and there were only two case where Ag could not be quantified in oysters. Of the 24 decreasing trends, only 8 do not include a zero Ag concentration at the end of the time series. A preponderance of zeroes also forced medians to be zero for

mussels in 1995 and 1996 and results for those years should be disregarded. That done, there is not a significant correlation between the annual median concentrations in mussels or oysters and year. Excluding 1995 and 1996 from the site-by-site analysis usually removed one year from the Ag data (since sites are visited biennially). More years were lost if samples from earlier years were unavailable for reanalysis (Daskalakis et al., 1997). In all, through 1994, there were 132 sites (rather than 196) with 6 or more years of data. Among those there were 15 decreasing trends for silver and 1 increase ($p \geq 0.05$). In summary there is a case, albeit weak, for an overall decreasing trend in Ag.

Of all the chemicals measured, chromium is the one with the lowest ratio of concentration in mollusk to concentration in sediment (Thomann et al., 1995). This means that it is the chemical most sensitive to the fact that mussels and oysters are not depurated after sampling. A small amount of entrained sediment will have a larger effect on measured Cr concentrations than on any other chemical. In and of itself, this would tend to make the Cr data noisy rather than create temporal trends. However, in recent years with the introduction of INAA Cr concentrations have increased. This is not because Cr has increased in mollusk tissue but because the Cr concentrations based on acid digests could not dissolve all the Cr in entrained sediment. It is known that Cr is incompletely and inconsistently extracted from sediment unless hydrofluoric acid is used (Willie and Berman, 1995). The chromium results in mollusks therefore are interpreted as an analytical artifact rather than as an increasing trend.

Discussion

Decreasing trends are good news but it is perhaps of equal importance that no chemical has been seen to be increasing in concentration. So, at least for the chemicals monitored by the National Status and Trends Program, contamination of the coastal United States is not increasing.

Decreasing trends in chemical concentrations are not unexpected. All the monitored chlorinated hydrocarbons have been banned for use in the United States. DDT was banned in 1972; PCB began being phased out in 1971 and further uses were banned in 1976; Dieldrin (and Aldrin) stopped being used in 1975. The agricultural use of chlordane was banned in 1983 and its use as a termiticide ended in 1988 (Shigenaka, 1990). In 1988, the U. S. Organotin Anti-Fouling Paint Control Act outlawed tributyltin application to boats of less than 25 m in length. Annual consumption of cadmium in the U.S. decreased over the period of 1986 through 1993 (O'Connor and Beliaeff,1995) and the decrease continued through 1996 (Kuck, 1997). There is no evidence that the use of copper, zinc, or silver has trended downwardin the United States. If concentrations of these elements are decreasing, the cause is probably increased care in the U.S. with regard to waste recycling and disposal.

Conclusion

The result of annually monitoring chemical concentrations in mussels and oysters in the coastal United States is the observation that the chlorinated hydrocarbons and tributyltin, chemicals banned for further use in the United States, are decreasing in concentration. There is a strong case that cadmium concentrations are decreasing in coastal waters in parallel with its decreased use in the United States. There are weak cases for decreases in copper, zinc, and silver. It is most noteworthy that none of the monitored chemicals are trending upwards in concentration.

References

Beliaeff, B., T. P. O'Connor, D. K. Daskalakis, and P. J. Smith. 1997. U. S. Mussel Watch Data from 1986 to 1994: Temporal Trend Detection at Large Spatial Scales. Environ. Sci. Tech. 31: 1411-1415

Capuzzo, J.M., J.W. Farrington, P. Rantamaki, C.H. Clifford, B.A. Lancaster, D.F. Leavitt and X. Jia, 1989. The relationship between lipid composition and seasonal differences in the distribution of PCBs in *Mytilus edulis* L. Mar. Environ. Res. 28: 259-264.

Cunningham, P.A. and M.R. Tripp, 1975. Factors affecting the accumulation and removal of mercury from tissues of the American oyster *Crassostrea virginica.* Mar. Biol. 31: 311-319.

Daskalakis, K. D., T. P. O'Connor, and E. A. Crecilius. 1997. Evaluation of Digestion Procedures for Determining Silver in Mussels and Oysters.Environ. Sci. Tech. 31: 2303-2306

Fisher, N.S. and J.-L. Teyssie, 1986. Influence of food composition on biokinetics and tissue distribution of zinc and americium in mussels. Mar. Ecol. Prog. Ser. 28: 197-207.

Keith, L. H. and W. A. Teillard. 1979. Priority pollutants I: a perspective view. Environ. Sci. Technol. 13: 416-423.

Kuck, P.H. 1997. Mineral commodity summaries. U.S. Geological Survey. Department of Interior, Washington, DC

Lauenstein, G. G. and A. Y. Cantillo (eds.), 1993a. Sampling and Analytical Methods of the NS&T Program National Benthic Surveillance and Mussel Watch Projects. Volume I. Overview and Summary of Methods. NOAA Technical Memorandum NOS ORCA 71 Silver Spring, MD.

Lauenstein, G. G. and A. Y. Cantillo (eds.), 1993b. Sampling and Analytical Methods of the NS&T Program National Benthic Surveillance and Mussel Watch Projects. Volume II. Comprehensive Descriptions of Complementary Measurements. NOAA Technical Memorandum NOS ORCA 71 Silver Spring, MD.

Lauenstein, G. G. and A. Y. Cantillo (eds.), 1993c. Sampling and Analytical Methods of the NS&T Program National Benthic Surveillance and Mussel Watch Projects. Volume III. Comprehensive Descriptions of Elemental Analytical Methods. NOAA Technical Memorandum NOS ORCA 71 Silver Spring, MD.

Lauenstein, G. G. and A. Y. Cantillo (eds.), 1993d. Sampling and Analytical Methods of the NS&T Program National Benthic Surveillance and Mussel Watch

Projects. Volume IV. Comprehensive Descriptions of Trace Organic Analytical Methods. NOAA Technical Memorandum NOS ORCA 71 Silver Spring, MD.

Lauenstein, G. G., A. Y. Cantillo, S. Kokkinakis, S. Frew, H. J. Jobling, and R. R. Fay. 1997. National Status and Trends Program: Mussel Watch Site Descriptions, through 1997. NOAA Technical Memorandum NOS ORCA 112 Silver Spring, MD.

Martin, M., 1985. State Mussel Watch: Toxics surveillance in California. Mar. Pollut. Bull. 16: 140-146.

Nriagu, J. O., 1989. A global assessment of natural sources of atmospheric trace elements. Nature 338: 47-49.

O'Connor, T. P., 1994. The National Oceanic and Atmospheric Administration (NOAA) National Status and Trends Mussel Watch Program: National Monitoring of Chemical Contamination in the Coastal United States. pp 331-349 In: C. R. Cothern and N. P. Ross (eds) *Environmental Statistics, Assessment, and Forecasting*. Lewis Publishers, Boca Raton, FL.

O'Connor, T. P. and B. Beliaeff. 1995. *Recent Trends in Coastal Environmental Quality: Results from the Mussel Watch Project*. NOAA, Silver Spring, MD 40 pp.

O'Connor, T. P. 1996. Trends in chemical concentrations in mussels and oysters collected along the U. S. coast from 1986 to 1993. Mar. Environ. Res. 41: 183-200

Phillips, D. J. H., 1980. *Quantitative Aquatic Biological Indicators*. Applied Science Publishers, London.

Phillips, D.J.H. and D.A. Segar, 1986. Use of bio-indicators in monitoring conservative contaminants: programme design imperatives. Mar. Pollut. Bull. 17: 10-17.

Pruell, R.J., J. G. Quinn, J. L. Lake, and W. R. Davis, 1987. Availability of PCBs and PAHs to *Mytilus edulis* from artificially resuspended sediments. pp 97-108 In Capuzzo, J. M. and D. R. Kester (eds.) *Oceanic Processes in Marine Pollution. Vol. 1. Biological Processes and Wastes in the Ocean*. Krieger, Malabar, FL.

Roesijadi, G., J. S. Young, A.S. Drum, and J. M. Gurtisen, 1984. Behavior of trace metals in *Mytilus edulis* during a reciprocal transplant field experiment. Mar. Ecol. Prog. Ser. 18: 155-170.

Sericano, J.L. 1993. The American oyster (*Crassostrea virginica*) as a bioindicator of trace organic contamination. Ph.D. Thesis Texas A&M University, College Station, TX 242 pp.

Shigenaka, G.,1990. Chlordane in the marine environment of the United States: Review and results from the National Status and Trends Program NOAA Technical Memorandum NOS ORCA 55 Seattle, WA .

Thomann, R. V., J. D. Mahoney, and R. Mueller. 1995. Steady-state model of biota sediment accumulation factor for metals in two marine bivalves. Environm. Toxicol. Chem. 14: 1989-1998

Wilkinson, L., 1987. The System for Statistics. SYSTAT Inc. Evanston, IL

Willie, S and S, Berman. 1995. NOAA National Status and Trends Program Fifth Round Intercomparison Exercise Results for Trace Metals in Marine Sediments and Biological Tissues. NOAA Technical Memorandum NOS ORCA 86, Silver Spring, MD 40 pp. & Appendices

Table 1. Chemicals measured in mollusks

Elements
Ag, Al, As, Cd, Cr, Cu, Fe, Hg, Mn, Ni, Pb, Se, Zn

Organic Compounds

Concentrations of individual compounds have been aggregated into groups.

∑PCB= twice the sum of concentrations of 18 congeners
PCB8, PCB18, PCB28, PCB44, PCB52, PCB66, PCB101, PCB105, PCB118, PCB128, PCB138, PCB153, PCB170, PCB180, PCB187, PCB195, PCB206, and PCB209

∑DDT= sum of concentrations of *ortho* and *para* forms of parent and metabolites
2,4'DDE; 4,4'DDE; 2,4'DDD; 4,4'DDD; 2,4'DDT; and 4,4'DDT

∑Cdane= sum of concentrations of four compounds
alpha-chlordane, *trans*-nonachlor, heptachlor, heptachlorepoxide

∑Dield= sum of concentrations of two compounds
aldrin and dieldrin

∑BT= sum of concentrations of parent compound and metabolites
monobutyltin, dibutyltin, and tributyltin [concentrations in terms of tin]

∑PAH= sum of concentrations of 24 compounds
naphthalene, 2-methylnaphthalene, 1-methylnaphthalene, biphenyl, 2,6-dimethylnaphthalene, acenaphthene, acenaphthylene, 1,6,7-trimethylnaphthalene, fluorene, phenanthrene, anthracene, 1-methylphenanthrene, fluoranthene, pyrene, benz[*a*]anthracene, chrysene, benzo[*b*]luoranthene, benzo[*k*]fluoranthene, benzo[*e*]pyrene, benzo[*a*]pyrene,perylene, dibenzanthracene, indeno[1,2,3-*cd*]pyrene, and benzo[*ghi*]perylene

Table 2. Lowest non-zero concentration, per chemical, in data sets used to identify trends. All concentrations given on a dry-weight basis

As	0.91	µg g^{-1}
Cd	0.071	
Cu	3.3	
Hg	0.0007	
Ni	0.15	
Pb	0.017	
Se	0.37	
Zn	55	
∑Cdane	0.033	ng g^{-1}
∑Dield	0.036	
∑DDT	0.51	
∑PCB	2.3	
∑PAH	2.6	
∑BT	0.27	ng of Sn g^{-1}

Table 3. Numbers of sites per chemical where there is a 95% level of confidence of increasing or decreasing trend in concentration in mollusks for 196 sites sampled in 6 or more years from 1986 through 1996. (Ag and Cr discussed separately, see text)

	Spearman		Kendall-tau	
Chem	Inc.	Dec.	Inc.	Dec.
As	13	11	10	12
Cd	0	26	0	25
Cu	8	19	6	23
Hg	9	13	7	13
Ni	8	7	6	7
Pb	15	12	15	10
Se	13	7	10	7
Zn	8	14	8	13
∑Cdane	1	83	1	73
∑DDT	0	40	0	35
∑Dield	1	30	1	28
∑PCB	3	29	3	23
∑PAH	9	10	6	10
∑BT	0	47	0	52
Ag	0	24	2	22
Cr	37	1	30	1

Table 4. Annual median concentrations (ppm-dry for trace elememts, ppb-dry for organic compounds), numb
(among sites with six or more years of data), Spearman coefficients for correlation between concentration an
>0.623 indicating correlation significant at $p<0.05$)

Year	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995
n	143	145	157	176	191	184	182	156	111	122
As	9.53	9.00	8.80	8.28	9.40	9.13	9.25	8.41	9.00	10.14
Cd	3.20	2.83	2.86	2.60	2.72	2.33	2.10	2.55	1.97	2.30
Hg	0.11	0.10	0.11	0.12	0.09	0.11	0.10	0.11	0.10	0.11
Ni	2.13	1.93	1.78	1.73	1.66	2.07	2.13	1.64	1.46	2.03
Se	2.57	2.60	2.90	2.27	2.41	2.58	2.58	2.48	2.59	3.25
Pb	0.75	0.86	0.74	0.63	0.78	0.77	0.72	0.85	1.06	0.74
Cu(oys)	103	114	138	118	134	118	120	117	94	134
Zn(oys)	1633	1759	2285	2246	2352	1948	2100	2086	2100	1982
Cu(muss)	9.80	9.97	9.73	9.93	8.59	8.83	8.67	8.37	8.67	8.76
Zn(muss)	143	133	128	120	133	130	120	130	120	120
∑CDANE	14.70	19.23	14.09	13.72	13.16	5.66	6.42	7.07	6.26	4.91
∑DIELD	5.79	8.30	4.50	4.21	3.28	3.02	3.64	3.61	2.87	2.70
∑DDT	37.07	39.58	37.40	35.54	30.19	18.55	24.78	24.59	26.50	23.26
∑PCB	145	121	136	118	110	60	72	62	103	72
∑PAH	.	.	503	270	251	233	241	245	228	192
∑BT	.	.	.	106.12	71.37	64.88	47.63	17.87	24.24	14.57

EXTRA References

Cantillo, A. Y. and R. Parris. 1993. Quality Assurance Project. Trace Organic Intercomparison Exercise Results. 1986-1990. NOAA Technical Memorandum NOS ORCA 69 Silver Spring, MD.

Cantillo, A. Y. 1994. Quality Assurance Project. Intercomparison Exercise Results 1991-1993. NOAA Technical Memorandum NOS ORCA 79 Silver Spring, MD.

Capuzzo, J.M., J.W. Farrington, P. Rantamaki, C.H. Clifford, B.A. Lancaster, D.F. Leavitt and X. Jia, 1989. The relationship between lipid composition and seasonal differences in the distribution of PCBs in *Mytilus edulis* L. Mar. Environ. Res. 28: 259-264.

Cunningham, P.A. and M.R. Tripp, 1975. Factors affecting the accumulation and removal of mercury from tissues of the American oyster *Crassostrea virginica.* Mar. Biol. 31: 311-319.

Department of Interior. 1994. Mineral Industry Surveys. Bureau of Mines Branch of Metals and Branch of Data Collection and Coordination. Washington, D.C.

Fisher, N.S. and J.-L. Teyssie, 1986. Influence of food composition on biokinetics and tissue distribution of zinc and americium in mussels. Mar. Ecol. Prog. Ser. 28: 197-207.

Helsel, D. R. and R. M. Hirsch, 1992. *Statistical Methods in Water Resources.* Elsevier, New York 522 pp.

Keith, L. H. and W. A. Teillard. 1979. Priority pollutants I: a perspective view. Environ. Sci. Technol. 13: 416-423.

Krahn, M. M., C. A. Wigren, R. W. Pearce, L. K. Moore, R. G. Bogar, W. D. MacLeod, Jr., S.-L. Chan and D. W. Brown, 1988. Standard analytical procedures for the NOAA National Analytical Facility, 1988: new HPLC cleanup and revised extraction procedures for organic contaminants. NOAA Technical Memorandum NMFS/NWC-153. NOAA/NMFS/NWFSC, Seattle, WA. 52 pp.

Lauenstein, G. G., M. F. Harmon, and B. W. Gottholm, 1993. National Status and Trends Program: Monitoring Site Descriptions (1984-1990) for the National Mussel Watch and Benthic Surveillance Projects. NOAA Technical Memorandum NOS ORCA 70 Silver Spring, MD.

Lauenstein, G. G. and A. Y. Cantillo (eds.), 1993a. Sampling and Analytical Methods of the NS&T Program National Benthic Surveillance and Mussel Watch Projects. Volume I. Overview and Summary of Methods. NOAA Technical Memorandum NOS ORCA 71 Silver Spring, MD.

Lauenstein, G. G. and A. Y. Cantillo (eds.), 1993b. Sampling and Analytical Methods of the NS&T Program National Benthic Surveillance and Mussel Watch Projects. Volume II. Comprehensive Descriptions of Complementary Measurements. NOAA Technical Memorandum NOS ORCA 71 Silver Spring, MD.

Lauenstein, G. G. and A. Y. Cantillo (eds.), 1993c. Sampling and Analytical Methods of the NS&T Program National Benthic Surveillance and Mussel Watch Projects. Volume III. Comprehensive Descriptions of Elemental Analytical Methods. NOAA Technical Memorandum NOS ORCA 71 Silver Spring, MD.

Lauenstein, G. G. and A. Y. Cantillo (eds.), 1993d. Sampling and Analytical Methods of the NS&T Program National Benthic Surveillance and Mussel Watch Projects. Volume IV. Comprehensive Descriptions of Trace Organic Analytical Methods. NOAA Technical Memorandum NOS ORCA 71 Silver Spring, MD.

Martin, M., 1985. State Mussel Watch: Toxics surveillance in California. Mar. Pollut. Bull. 16: 140-146.

Nriagu, J. O., 1989. A global assessment of natural sources of atmospheric trace elements. Nature 338: 47-49.

O'Connor, T.P., 1992. *Mussel Watch*: *Recent Trends in Coastal Environmental Quality* NOAA Rockville, MD. 46 pp.

O'Connor, T. P., 1994. The National Oceanic and Atmospheric Administration (NOAA) National Status and Trends Mussel Watch Program: National Monitoring of Chemical Contamination in the Coastal United States. pp 331-349 In: C. R. Cothern and N. P. Ross (eds) *Environmental Statistics, Assessment, and Forecasting*. Lewis Publishers, Boca Raton, FL.

O'Connor, T. P., A. Y. Cantillo, and G. G. Lauenstein,1994. Monitoring of Temporal Trends in Chemical Contamination by the NOAA National Status and Trends Mussel Watch Project. pp 29-50 In: K. J. M. Kramer (ed) *Biomonitoring of Coastal Waters and Estuaries*. CRC Press, Boca Raton, FL.

Phillips, D. J. H., 1980. *Quantitative Aquatic Biological Indicators*. Applied Science Publishers, London.

Phillips, D.J.H. and D.A. Segar, 1986. Use of bio-indicators in monitoring conservative contaminants: programme design imperatives. Mar. Pollut. Bull. 17: 10-17.

Pruell, R.J., J. G. Quinn, J. L. Lake, and W. R. Davis, 1987. Availability of PCBs and PAHs to *Mytilus edulis* from artificially resuspended sediments. pp 97-108 In Capuzzo, J. M. and D. R. Kester (eds.) *Oceanic Processes in Marine Pollution. Vol. 1. Biological Processes and Wastes in the Ocean*. Krieger, Malabar, FL.

Roesijadi, G., J. S. Young, A.S. Drum, and J. M. Gurtisen, 1984. Behavior of trace metals in *Mytilus edulis* during a reciprocal transplant field experiment. Mar. Ecol. Prog. Ser. 18: 155-170.

Seed, R., 1992. Systematics evolution and distribution of mussels belonging to the genus *Mytilus*: an overview. Amer. Malac. Bull. 9:123-137.

Sericano, J.L.,1993. The American oyster (*Crassostrea virginica*) as a bioindicator of trace organic contamination. Ph.D. Thesis Texas A&M University, College Station, TX 242 pp.

Shigenaka, G.,1990. Chlordane in the marine environment of the United States: Review and results from the National Status and Trends Program NOAA Technical Memorandum NOS ORCA 55 Seattle, WA .

Valette-Silver, N., 1992. Elemental Analyses of Marine Sedimemts and Biological Tissues. NOAA Technical Memorandum NOS ORCA 66 Silver Spring, MD.

Wilkinson, L., 1987. The System for Statistics. SYSTAT Inc. Evanston, IL

Zar, J. H., 1984. *Biostatistical Analysis*. Second Edition. Prentice-Hall, Englewood Cliffs, NJ 718 pp.

Possibe references

Daskalakis, K. D. and T. P. O'Connor 1995. Distribution of chemicalcontamination in coastal and estuarine sediments. *Mar. Environ. Res.* 40: 381-398

O'Connor, T. P. and B. Beliaeff. 1995. *Recent Trends in Coastal Environmental Quality: Results from the Mussel Watch Project*. NOAA, Silver Spring, MD 40 pp.

O'Connor, T. P. 1996. Trends in chemical concentrations in mussels and oysters collected along the U. S. coast from 1986 to 1993. *Mar. Environ. Res.* 41: 183-200

O'Connor, T. P. 1996. Coastal Sediment Contamination in the Northeast Shelf Large Marine Ecosystem. pp. 239-257 In: K. Sherman, N. A. Jaworsky, and T. J. Smayda (eds.) *The Northeast Shelf Ecosystem: Assessment, Sustainability, and Management*. Blackwell Science, Cambridge MA

Beliaeff, B., T. P. O'Connor, D. K. Daskalakis, and P. J. Smith. 1997. U. S. Mussel Watch Data from 1986 to 1994: Temporal Trend Detection at Large Spatial Scales. *Environ. Sci. Tech.* 31: 1411-1415

Daskalakis, K. D., T. P. O'Connor, and E. A. Crecilius. 1997. Evaluation of Digestion Procedures for Determining Silver in Mussels and Oysters.*Environ. Sci. Tech.* 31: 2303-2306

Beliaeff, B., T. P. O'Connor, D. D. Claisse. 1997. Comparison of Chemical Concentrations in Mussels and Oysters from the United States and France. *Environm. Monit. and Assess.* in press

O'Connor, T. P., K. D. Daskalakis, J. L. Hyland, J. F. Paul. and J. K.Summers. 1997. Comparisons of measured sediment toxicity with predictions based on chemical guidelines. *Environ. Toxicol. Chem.* In press

Thomann, R. V., J. D. Mahoney, and R. Mueller. 1995. Steady-state model of biota sediment accumulation factor for metals in two marine bivalves. Environm. Toxicol. Chem. 14: 1989-1998

Willie, S and S, Berman. 1995. NOAA National Status and Trends Program Fifth Round Intercomparison Exercise Results for Trace Metals in Marine Sediments and Biological Tissues. NOAA Technical Memorandum NOS ORCA 86, Silver Spring, MD 40 pp. & Appendices